Time-of-Flight Neutron Reflectometry

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Materials Science Division

Instrument Scientist of POSYI reflectometer

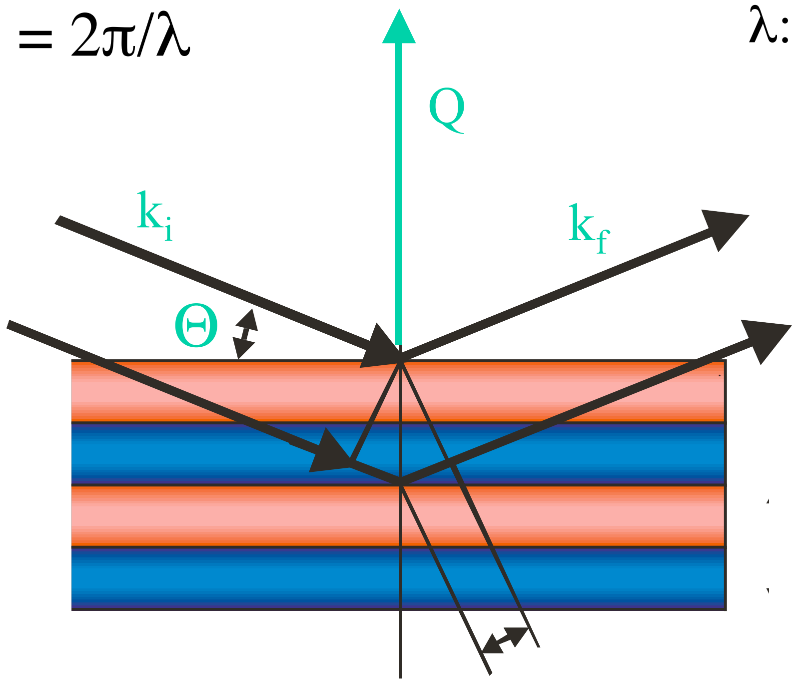
A) **Introduction:**
   Time of Flight Neutron Reflectometry

B) **Your Experiment:**
   Neutron Reflectivity from Polymer Films on Si

Neutron Reflectivity

Θ: angle of incidence
λ: wavelength

$|\mathbf{k}| = 2\pi/\lambda$



The reflectivity of the sample is measured as a function of the scattering vector Q

$$\mathbf{Q} = -\mathbf{k}_i + \mathbf{k}_f$$
$$|\mathbf{Q}| = 4\,\pi \sin\Theta / \lambda$$

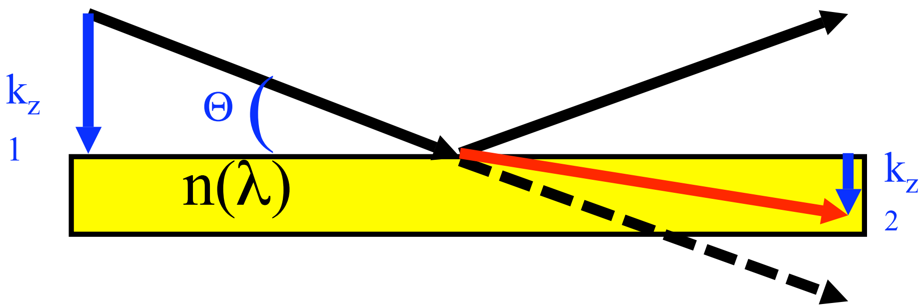
=> two concepts for neutron reflectivity measurements:

a) fixed wavelength + variable angle

b) variable wavelength + fixed angle

Total Reflection at Surfaces

$n_{vac}=1$



For neutrons (and X-rays) with wavelengths of a few Å, almost all materials have an optical index slightly smaller than 1.

=> Total reflection up to a critical angle $\Theta_{crit}(\lambda)$

Refraction index:

$n(\lambda) = k_{z2}$ **(inside the media) /** k_{z1} **(outside)**

Kinetic energy of a free particle:

$E_1 = \ddot{y}^2 k_{z1}^2 / 2m_N$

Inside the media with potential V, k_{z2} is (in most cases) smaller (conservation of energy):

$\ddot{y}^2 k_{z2}^2 / 2m_N + V = E_1$

$\Rightarrow \quad k_{z2} = (k_{z1}^2 - 2m_N V / \ddot{y}^2)^{1/2}$

Connection to microscopic properties:

Fermi pseudo potential: $V = 2\pi \ddot{y}^2 N b / m_N$

with N: number density [at/cm^3]
b: coherent scattering length of the nuclei in the material [fm]

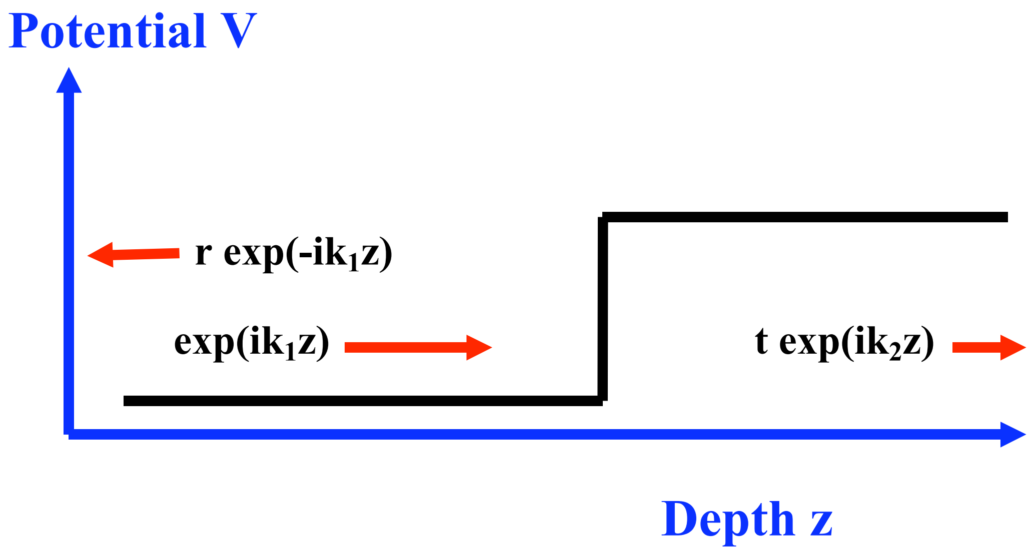
Critical angle for total reflection is reached, if $E_z = V$ **!**

$\Theta_{crit} = \sin^{-1}\lambda(N \cdot b/\pi)^{1/2} = \cos^{-1} n$

or

$Q_{crit} = 4\pi \sin\Theta / \lambda = 4(\pi N \cdot b)^{1/2}$

Calculation of the reflectivity at a potential step



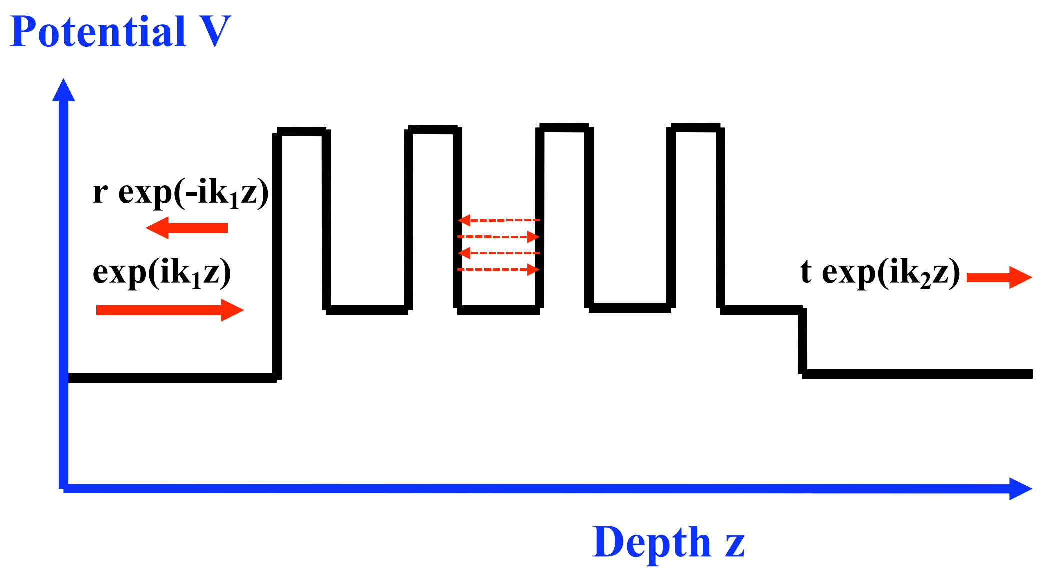
Solution of the quantum mechanic problem:

Fresnel equations

Reflectivity $R = |\, r \,|^2 = |\, (k_1 - k_2) / (k_1+k_2) \exp(i2k_1z) \,|^2$

Transmission $T = |\, t \,|^2 = |\, 2k_1 / (k_1+k_2) \exp(i2(k_1 - k_2)z) \,|^2$

Example: Potential of a multilayer

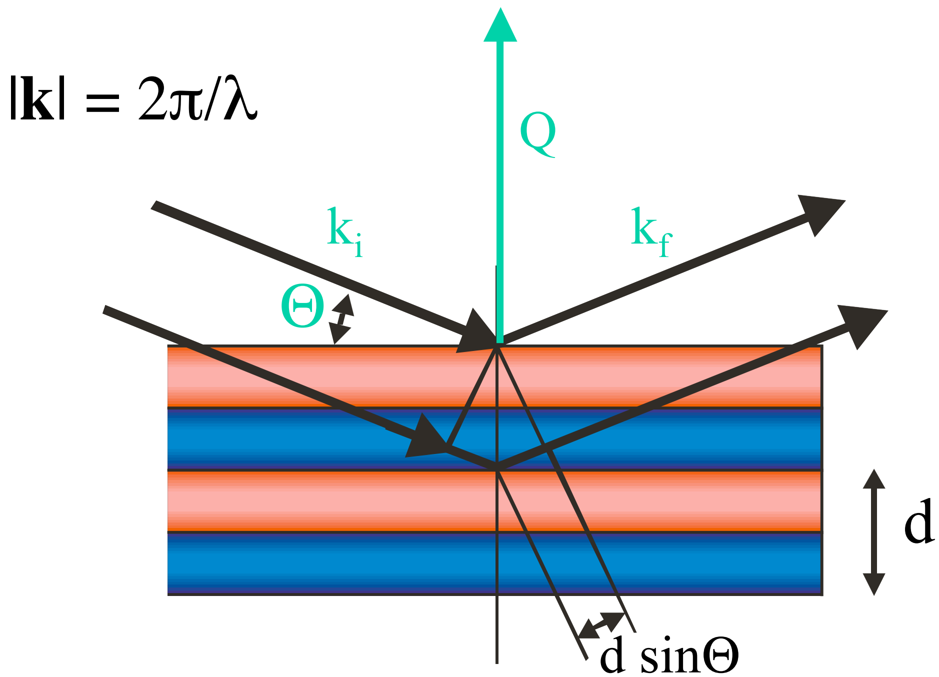


At each interface one has to take into account:

- **Refraction effects**
- **Multiple-scattering effects**

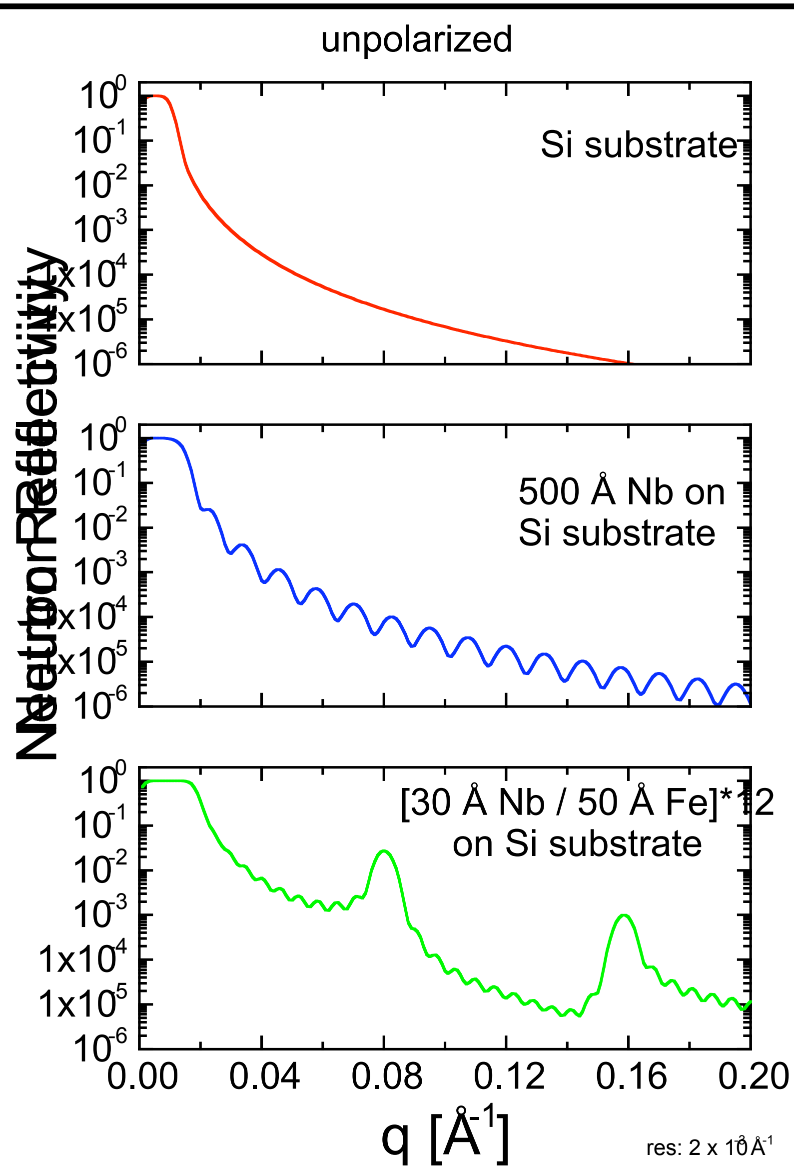
Bragg's Law for Periodic Layered Structures

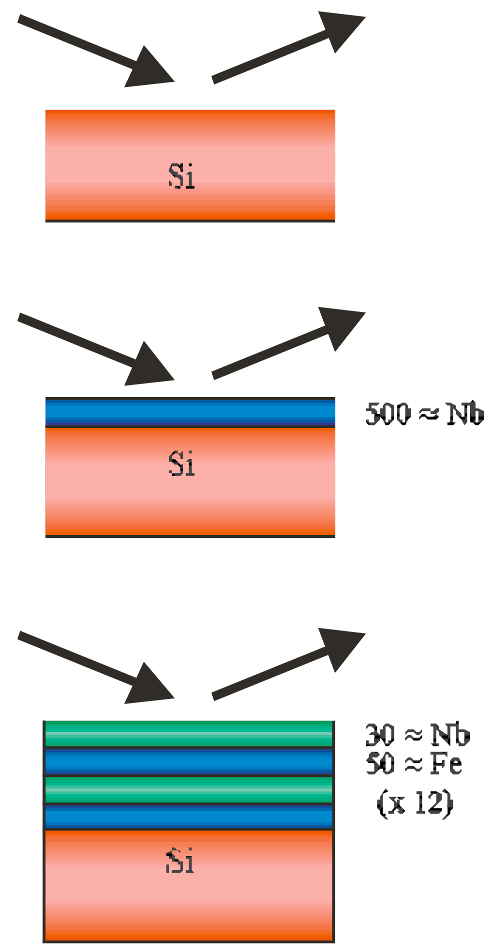
constructive interference if: $2d \sin\Theta = n \lambda$



d: double layer thickness
Θ: angle of incidence
n: order number (0,1,2,...)
λ: wavelength

Reflectivity of Layered Structures

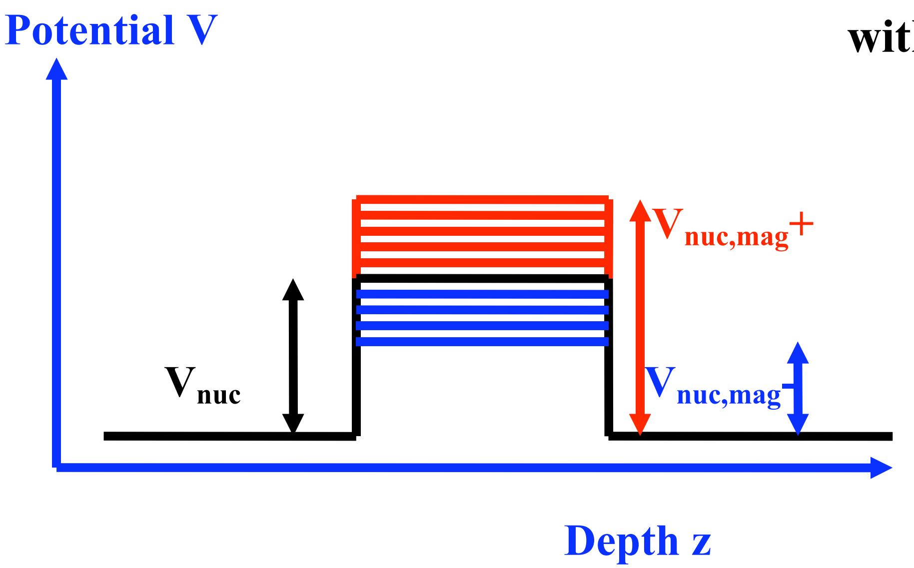




Reflectivity of Magnetic Layers

Fermi pseudo potential:

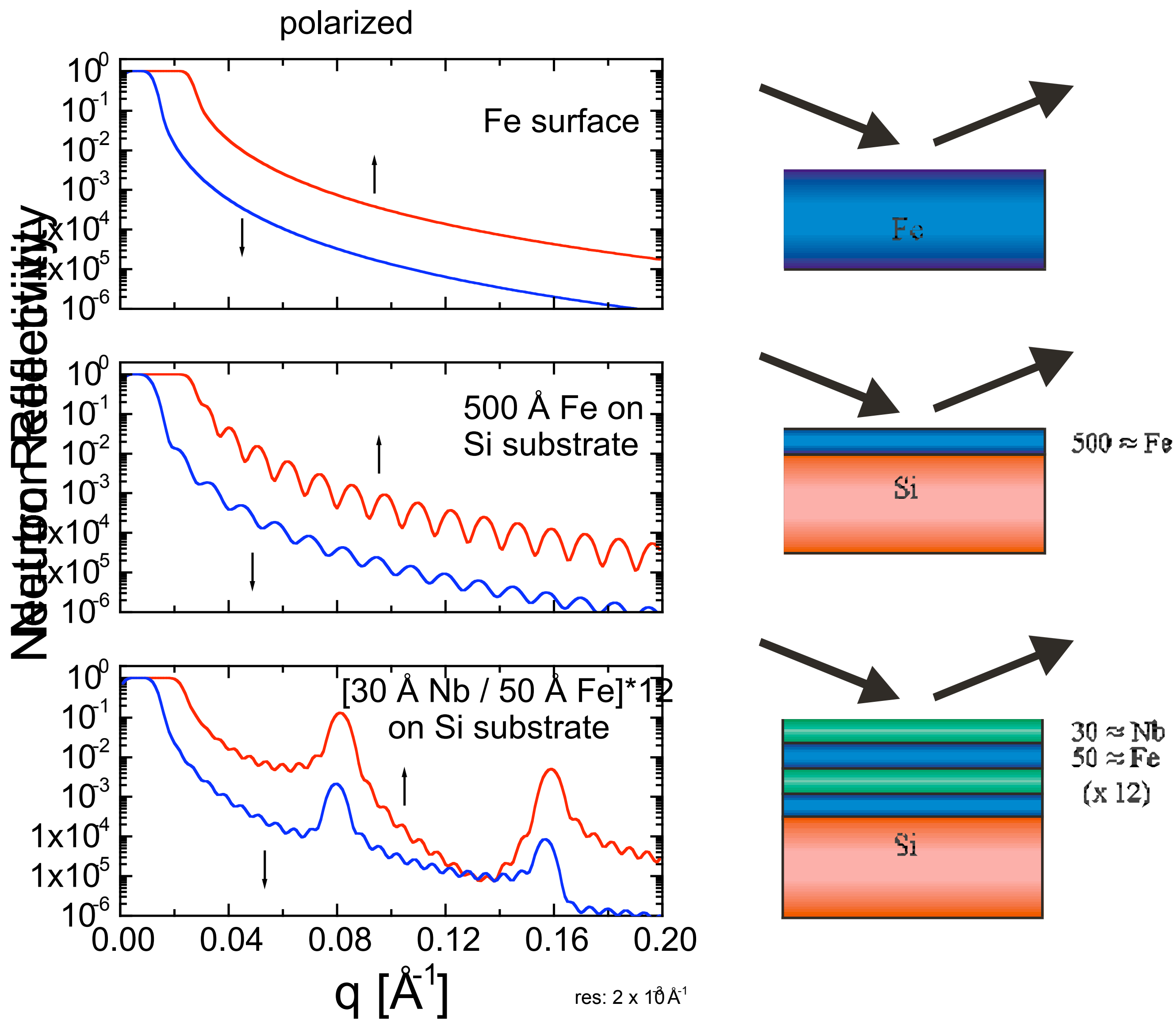
$$V = 2\pi \hbar^2 N (b_n +/- b_{mag})/m_N$$



with b_{nuc}: nuclear scattering length [fm]
b_{mag}: magnetic scattering length [fm]
(1 μ_B/Atom => 2.695 fm)
N: number density [at/cm^3]
m_N: neutron mass

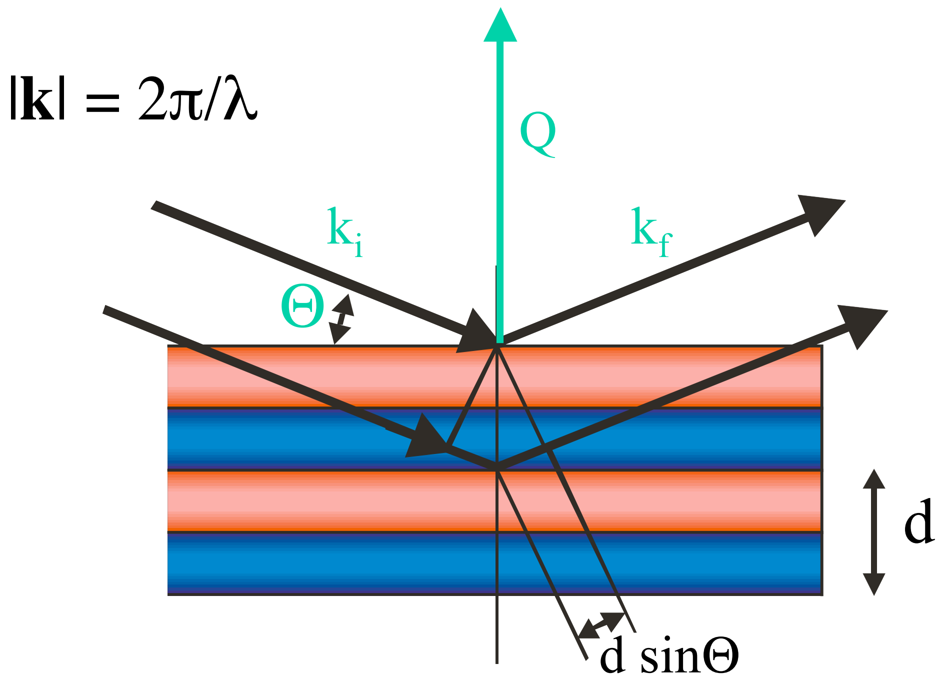
Spin"up" neutrons see a high potential.
Spin"down" neutrons see a low potential.

Polarized Neutron Reflectivity of Layered Magnetic Structures



Bragg's Law for Periodic Layered Structures

constructive interference if: $2d \sin\Theta = n \lambda$



d: double layer thickness
Θ: angle of incidence
n: order number (0,1,2,...)
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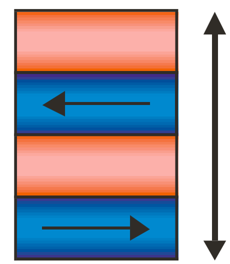
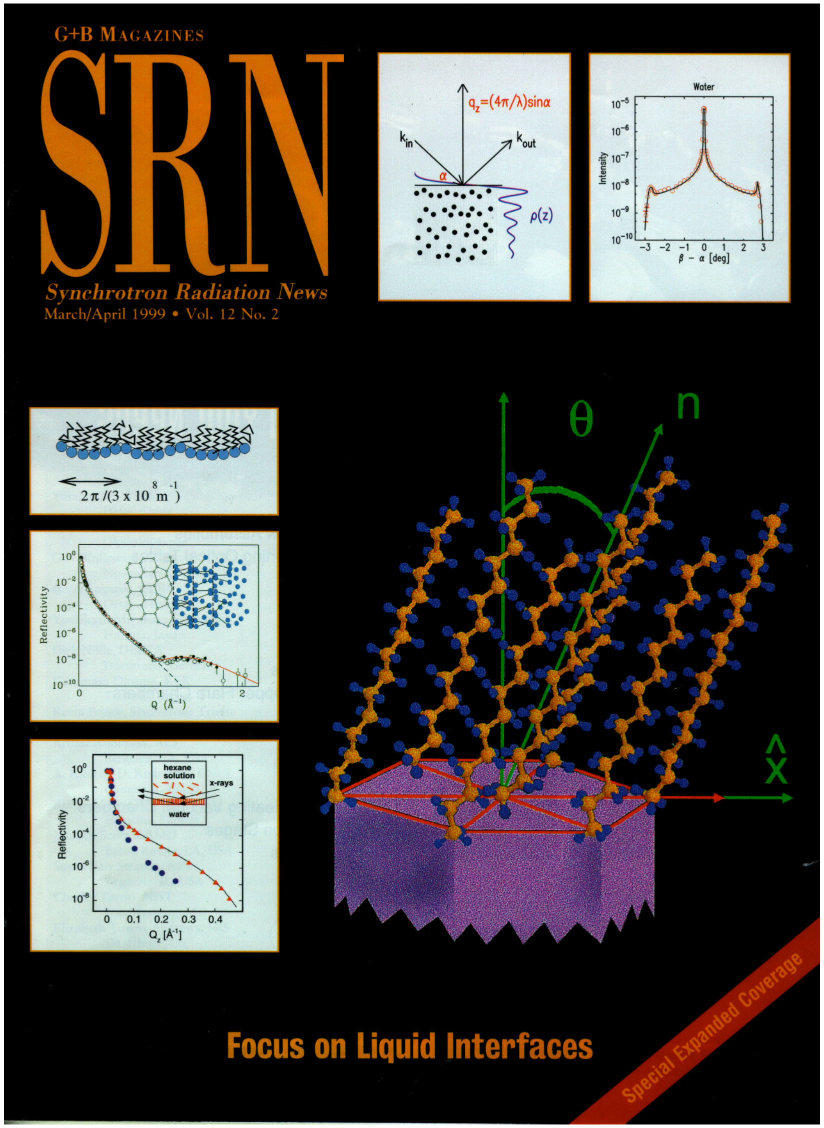


d_{mag}: magnetic periodicity

example:
antiferromagnetic coupling of magnetic layers

Experiments with “Soft” Matter and Liquids



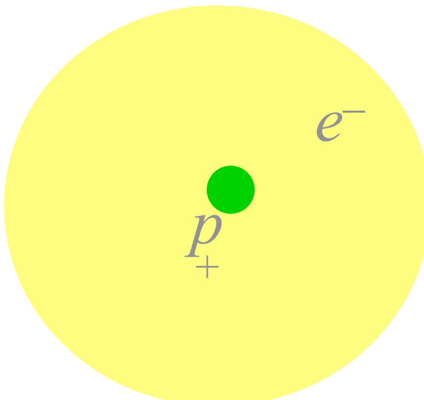
Research topics:

- morphology and thermodynamics of ultrathin polymer films
- monolayer films on liquids
- liquid/liquid and liquid/solid interfaces
- structure evolution
- diluted systems

- much more !!!

Why are (rare) Neutrons an excellent Probe for Soft Matter?

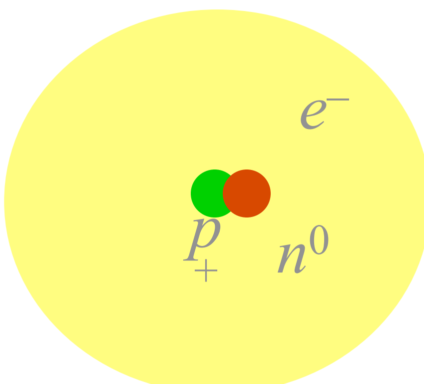
Hydrogen and Deuterium Labeling



Hydrogen

$$b_{xray} = 0.282 \times 10^{-4} \text{ Å}$$

$$b_{neutron} = -0.374 \times 10^{-4} \text{ Å}$$



Deuterium

$$b_{xray} = 0.282 \times 10^{-4} \text{ Å}$$

$$b_{neutron} = 0.665 \times 10^{-4} \text{ Å}$$

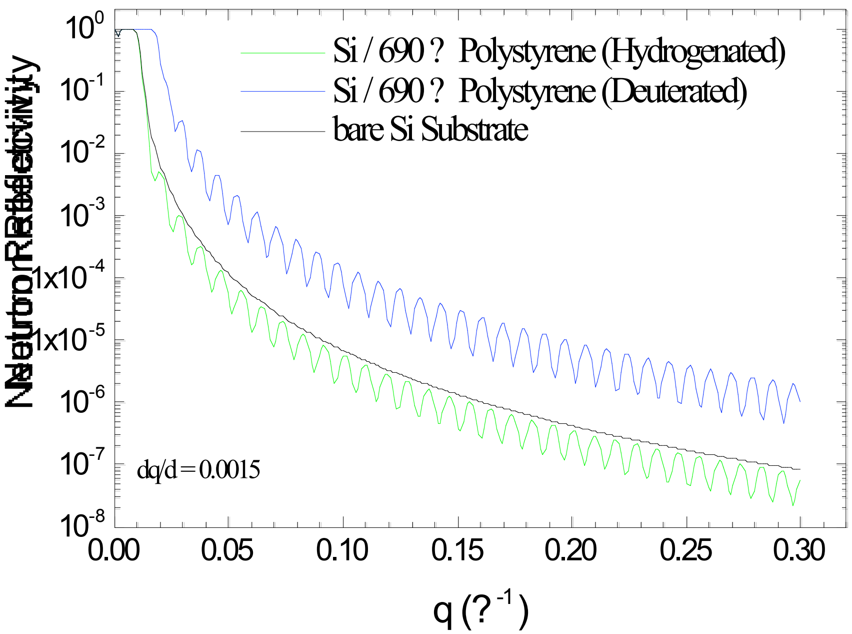
Hydrogen and deuterium have a huge difference in the interaction strength with neutrons !

Examples for Hydrogen/Deuterium Contrast Variation

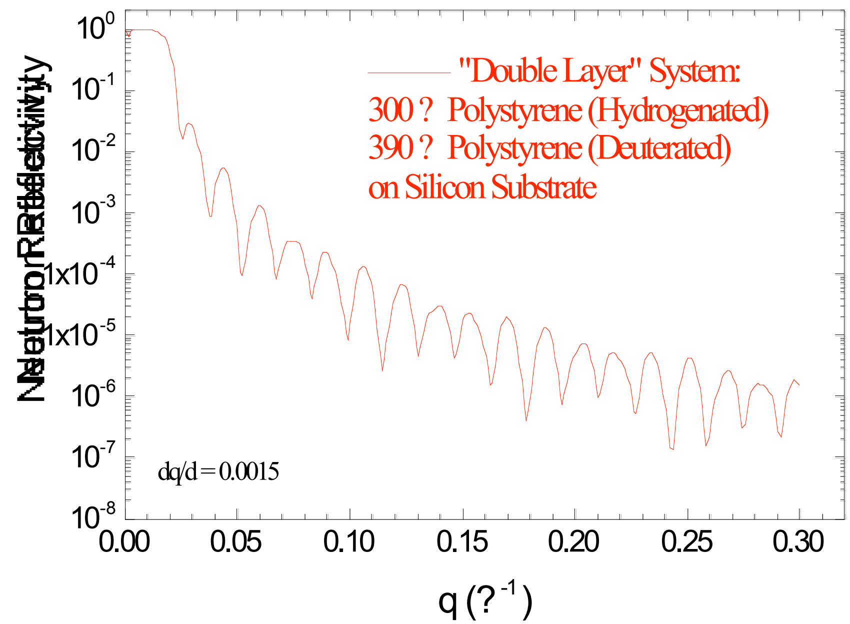
Neutron Scattering Length Density for Polystyrene

hydrogenated (C_8H_8): 1.4×10^{-6} Å^{-1}

deuterated (C_8D_8): 6.4×10^{-6} Å^{-1}



dPS has a much higher edge of total reflectivity !



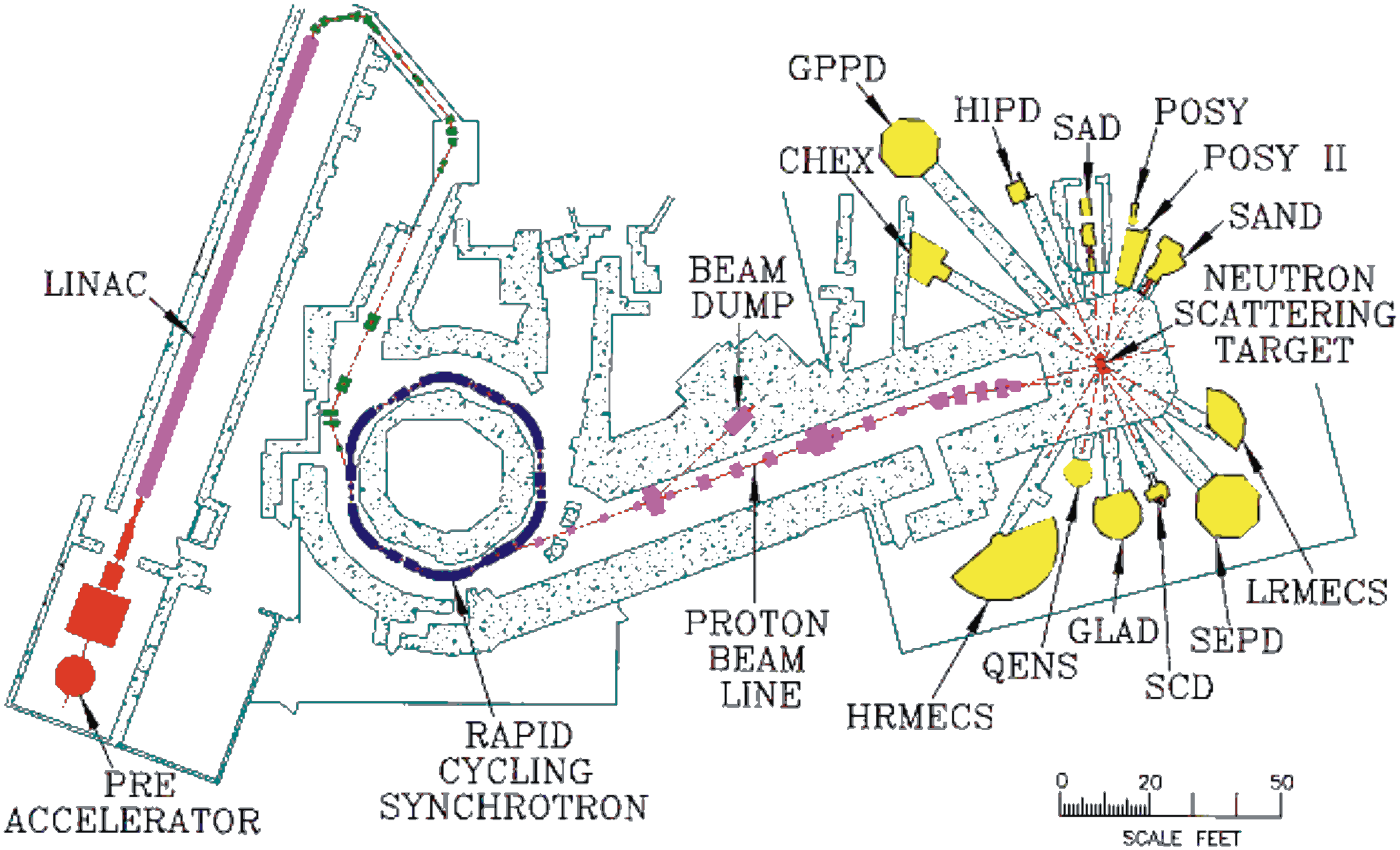
"Double layer" system shows two periodicities:

a) low frequency contribution from 390 Å dPS

b) high frequency contribution from 690 Å total film thickness (barely visible)

The oscillations from 390 Å dPS dominate due to its high potential !

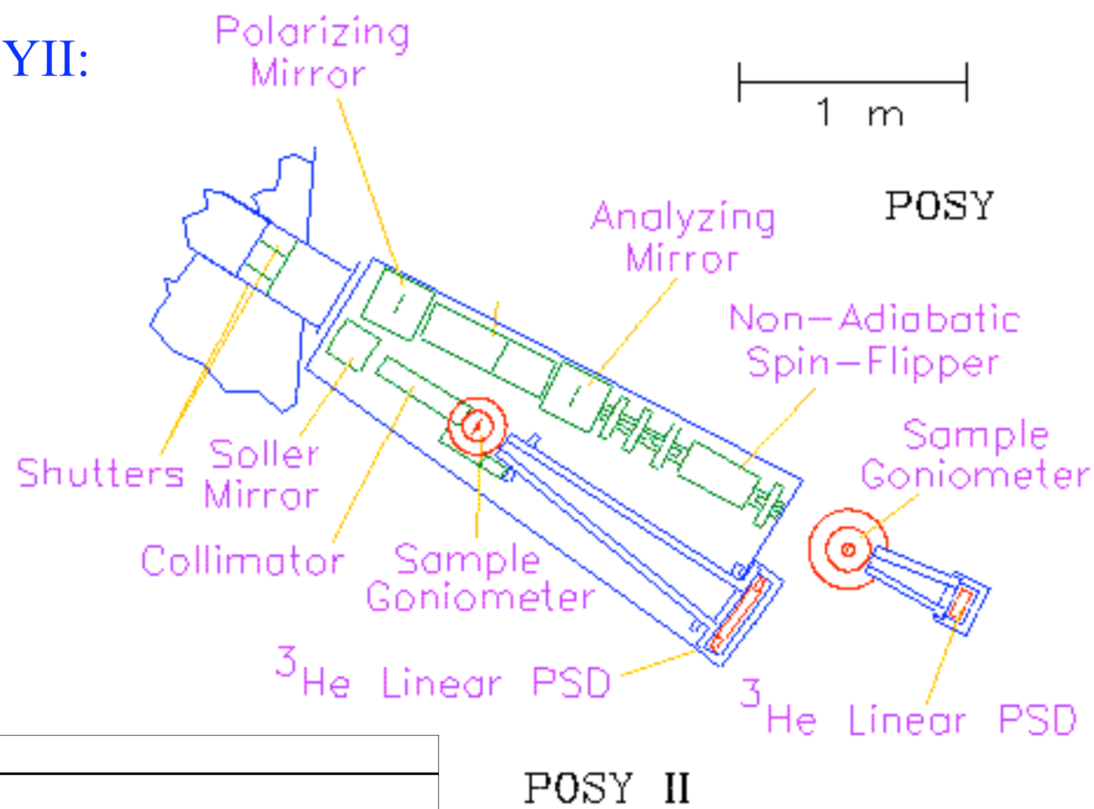
The Intense Pulsed Neutron Source Facilities



Neutrons are created by a 450 MeV/14 μA (=6.3 kW) proton beam which hits a Uranium target. The protons strike the target in 80 ns long pulses with a frequency of 30 Hz. The Uranium "boils off" neutrons in spallation and fast fission processes. The fast neutrons emerging the target are slowed down by a liquid hydrogen moderator ($T_{eff.}$= 32 K).

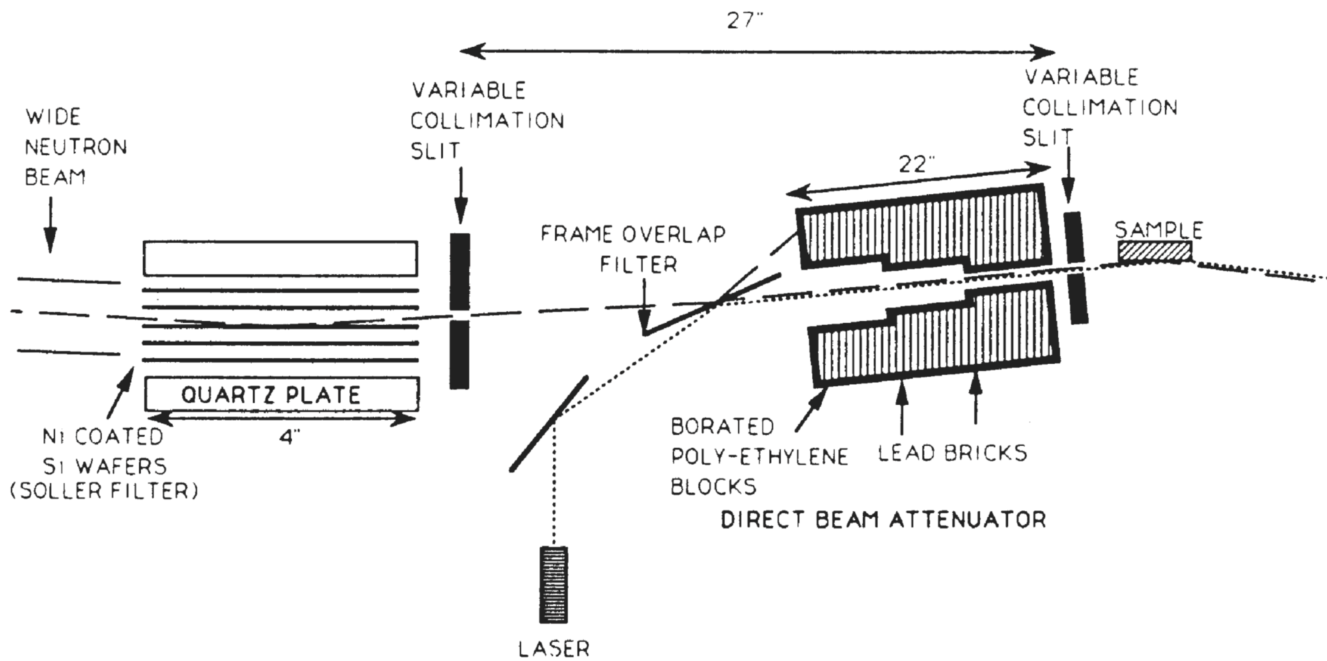
The IPNS Neutron Reflectometer POSY II

Instrument Scientist POSYII:
Rick Goyette



Beam Line	C2
Initial Flight Path	6.2 m
Final Flight Path	1.8 m
Beam Size	50 mm x (0 -3 mm)
Detector	Linear Position Sensitive Detector 20 cm
Choppers	none
Intensity	100 neutrons/pulse
Wave-vector Range	0-0.25 Å^{-1}
Wave-vector Resolution	1 x 10^{-4} Å^{-1}

The Filter/Collimation System of POSY II



Soller filter:	reflects λ > 2.5 Å to the sample
Frame overlap filter:	reflects λ > 16 Å out of the beam

=>

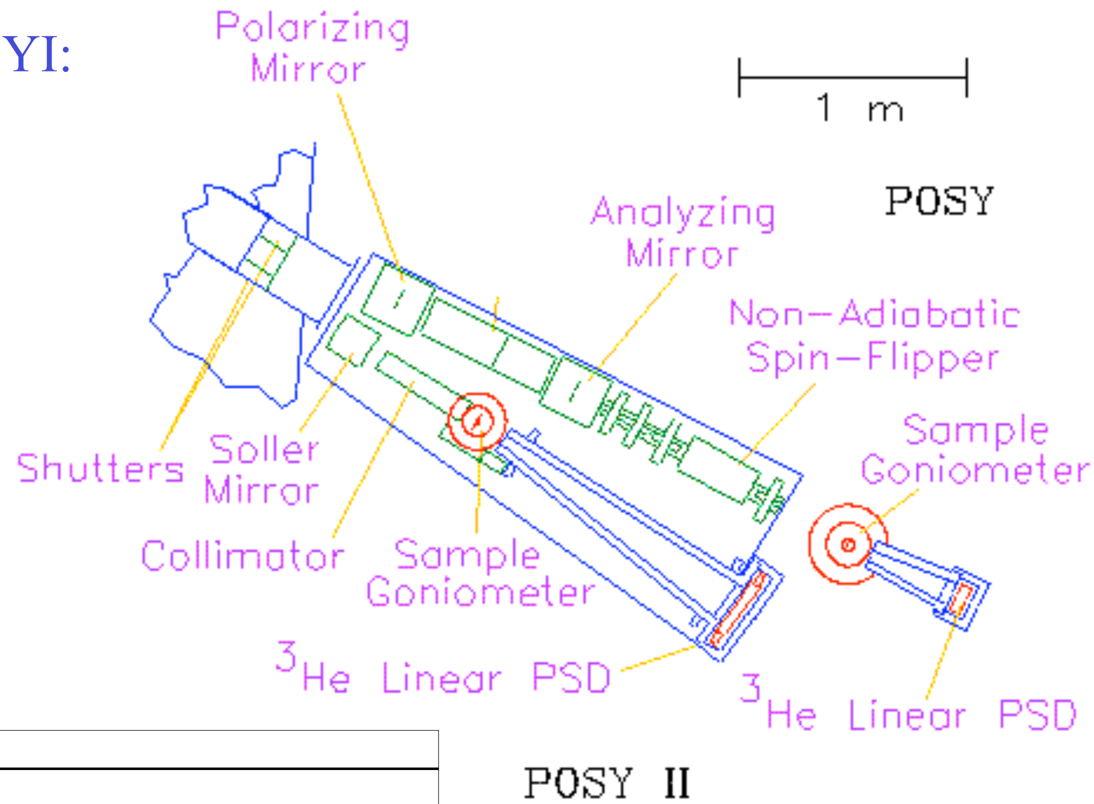
Neutron spectrum of POSY II: 2.5 Å > λ > 16 Å

The IPNS Neutron Reflectometer POSY I

Instrument Scientist POSYI:
Suzanne te Velthuis

Scientific Associate:
Rick Goyette

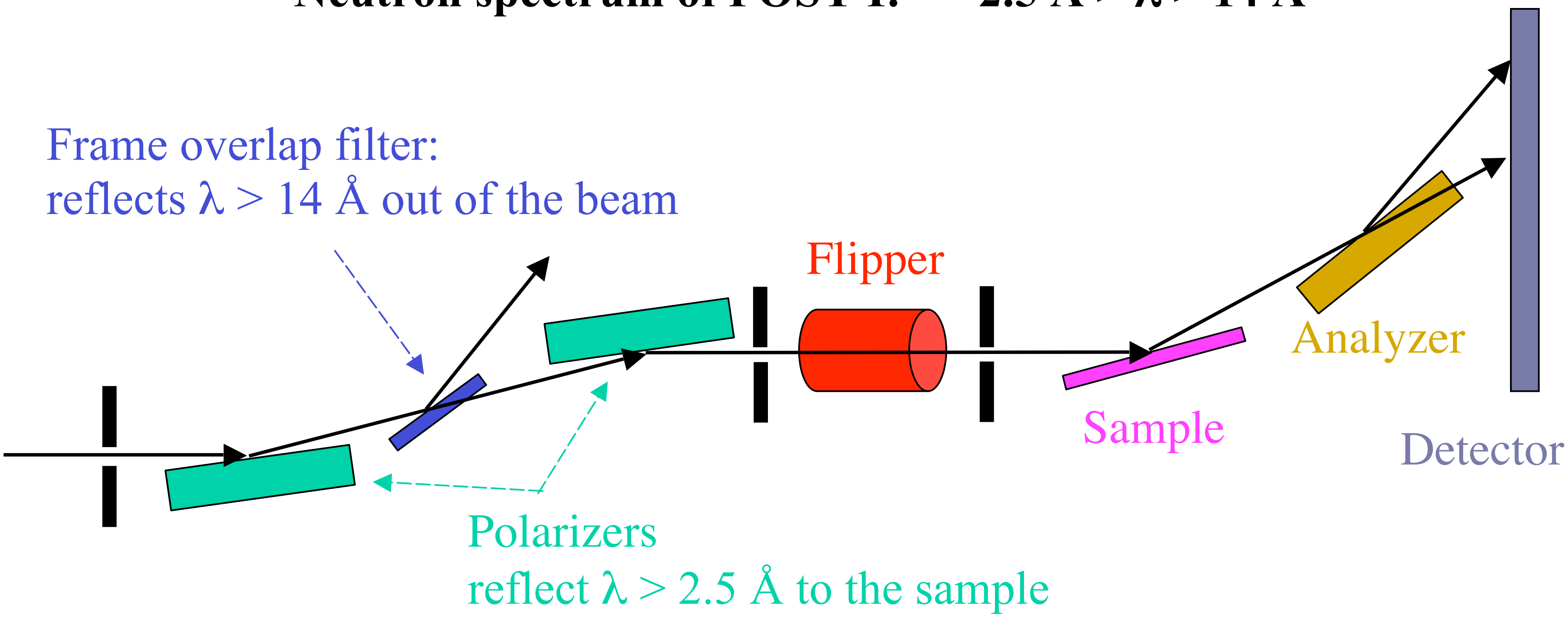
Post Doc:
Abdel Al-Smadi

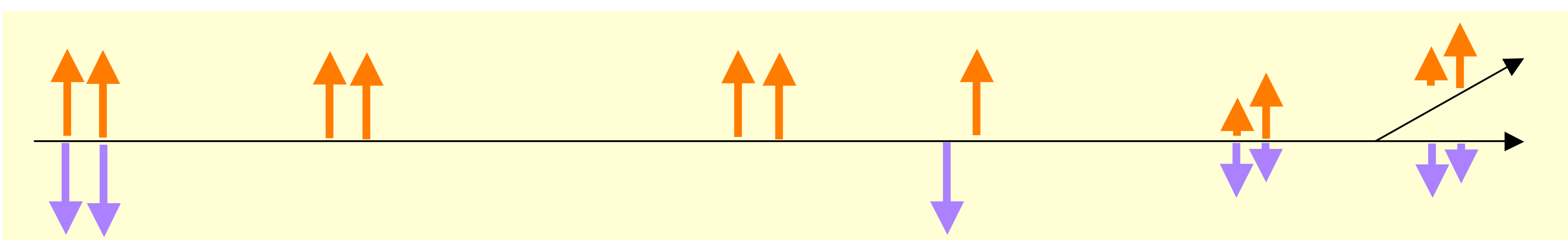


Beam Line	C2
Initial Flight Path	8.3 m
Final Flight Path	0.9 m
Beam Size	(0-0.3) x 25mm
Detector	Linear Position Sensitive Detector 20 cm
Choppers	none
Intensity	40 neutrons/pulse
Wave-vector Range	0- 0.08 Å^{-1}
Wave-vector Resolution	2 x 10^{-4} Å^{-1}

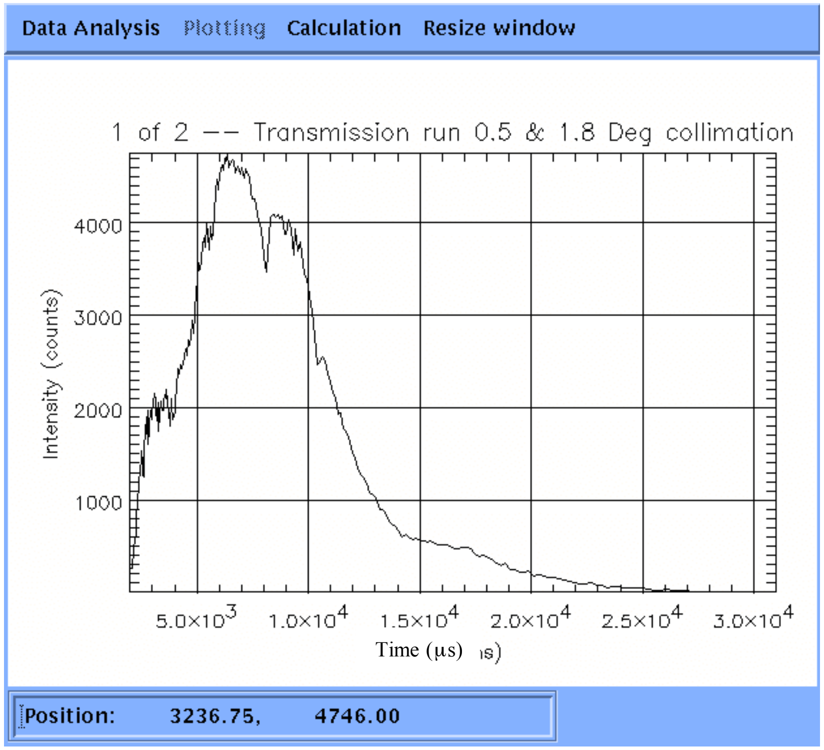
The Filter/Collimation System of POSY I

Neutron spectrum of POSY I: 2.5 Å > λ > 14 Å





Time-of-flight Spectrum at Detector Position



Relation between wavelength λ and time of flight t_{TOF}:

$$\lambda = \frac{h}{m_n L_{TOF}} \cdot t_{TOF}$$

with:

Planck's constant $h = 6.626 \cdot 10^{-34}$ Js

neutron mass $m_n = 1.675 \cdot 10^{-27}$ kg

distance source/detector $L_{TOF} = 7.750$ m

$\lambda = 2.5$ Å => $t_{TOF} = 4898$ μs

$\lambda = 16$ Å => $t_{TOF} = 31348$ μs

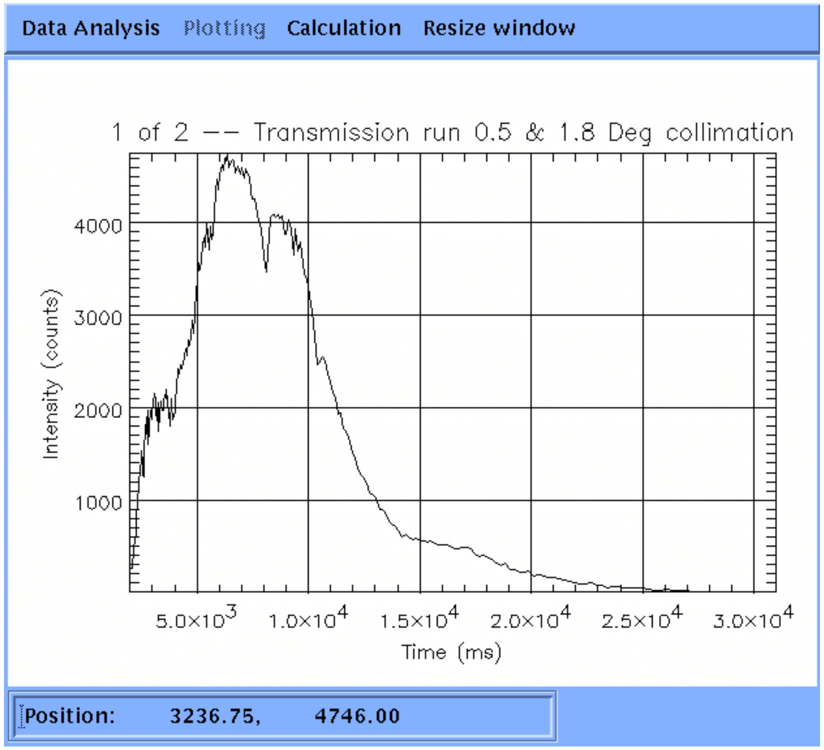
new pulse starts at: 33333 μs

λ (Å) $= 5.104 \cdot 10^{-4} \cdot t_{TOF}$ (μs)

Performing a Time-of-Flight Neutron Reflectometry Experiment

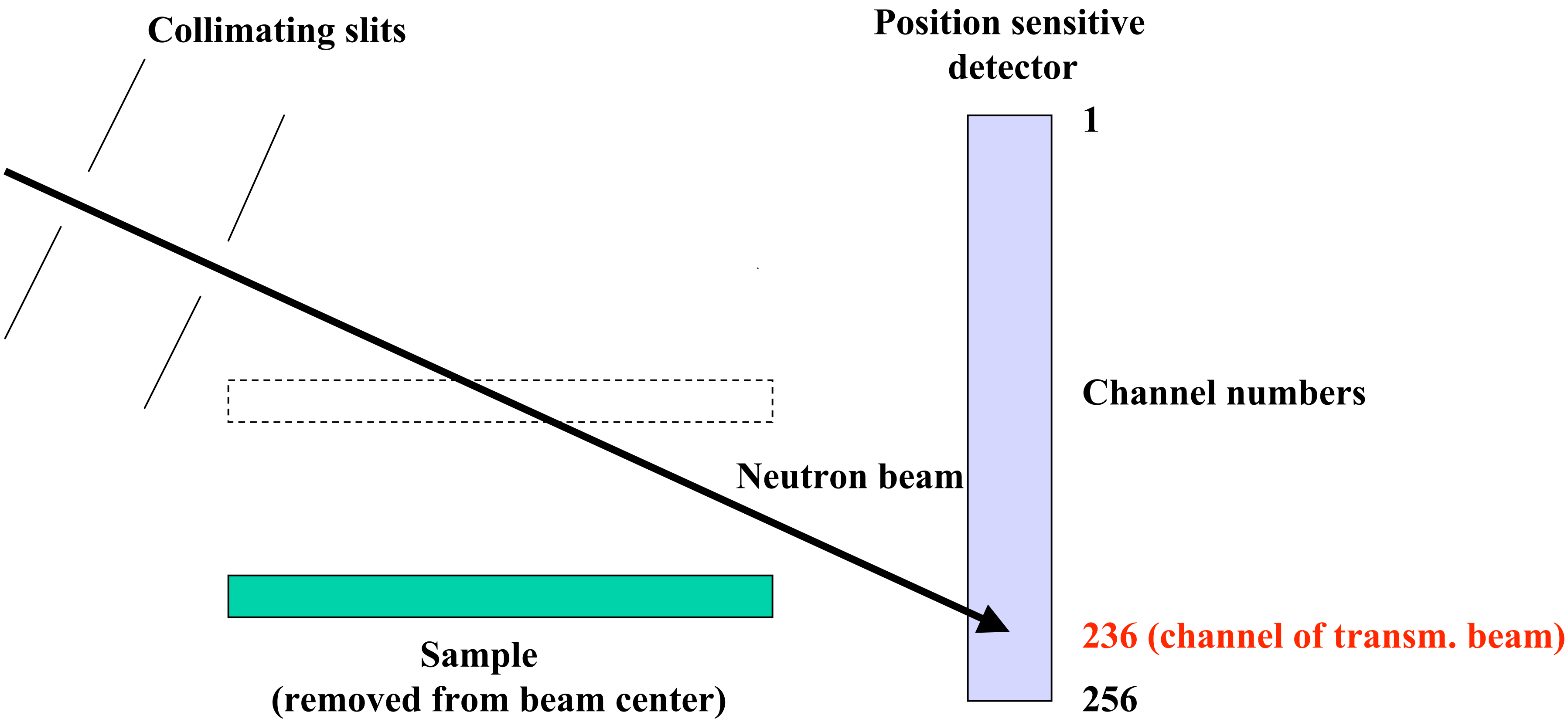
1. Step:
Measure a “Transmission Run”

(This is a measurement without a sample in the beam in order to determine the incident wavelength spectrum.)

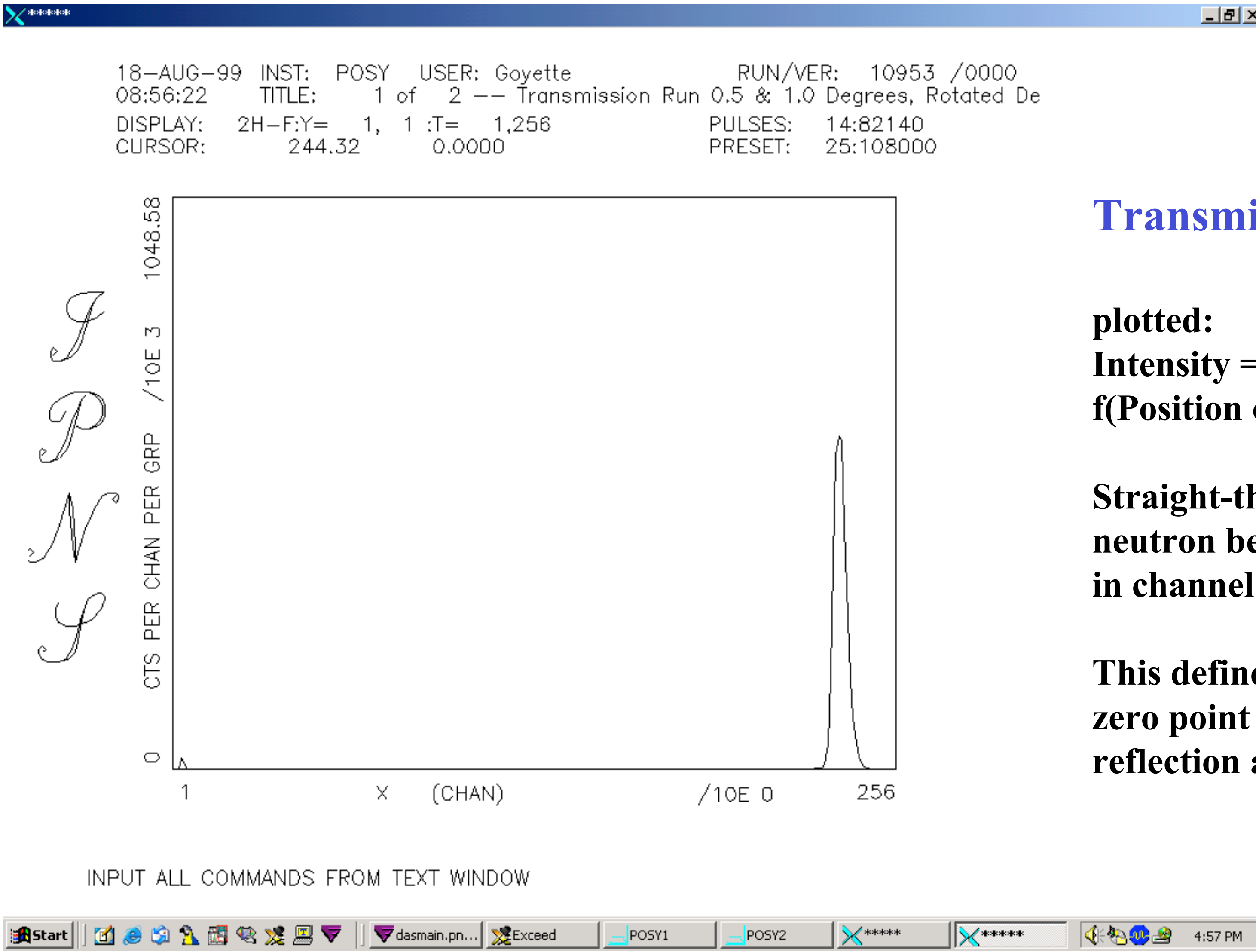


Looking at the Raw Data (POSYII)

Setup for a transmission run:
(top view)



Looking at the Raw Data (POSYII)



Transmission run:

plotted:
Intensity =
f(Position on detector)

Straight-through neutron beam is in channel 236.

This defines the zero point of the reflection angle.

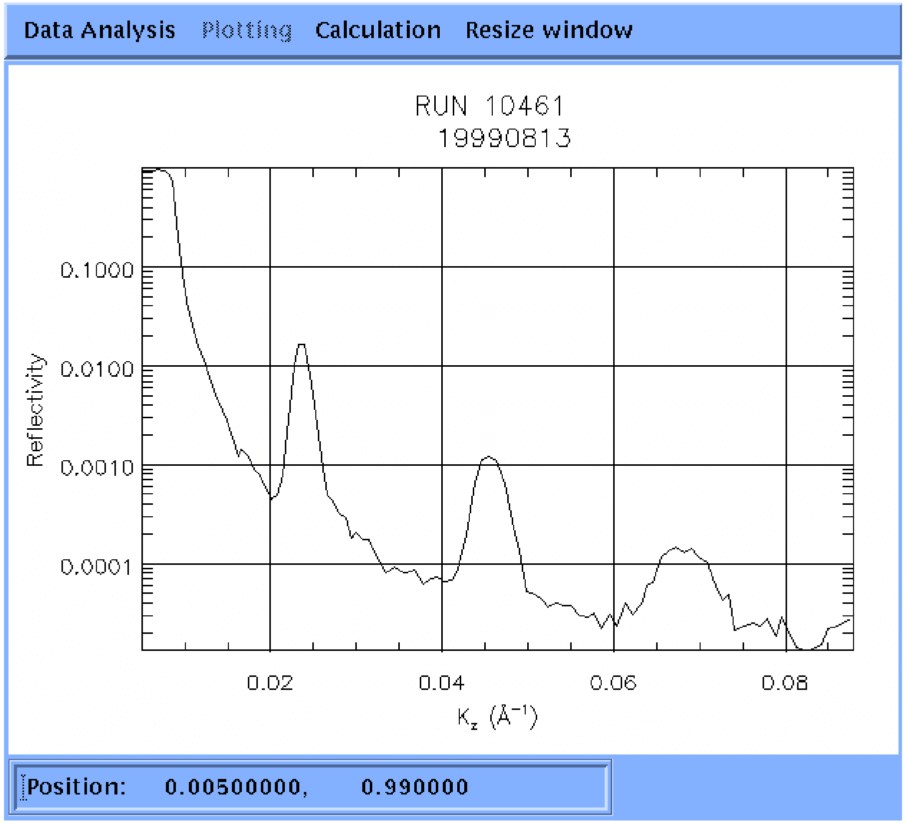
Performing a Time-of-Flight Neutron Reflectometry Experiment

2. Step:
Insert the sample and measure a "Reflectivity Run"

(Choose the scattering angle appropriately such that the run covers the desired Q range.)

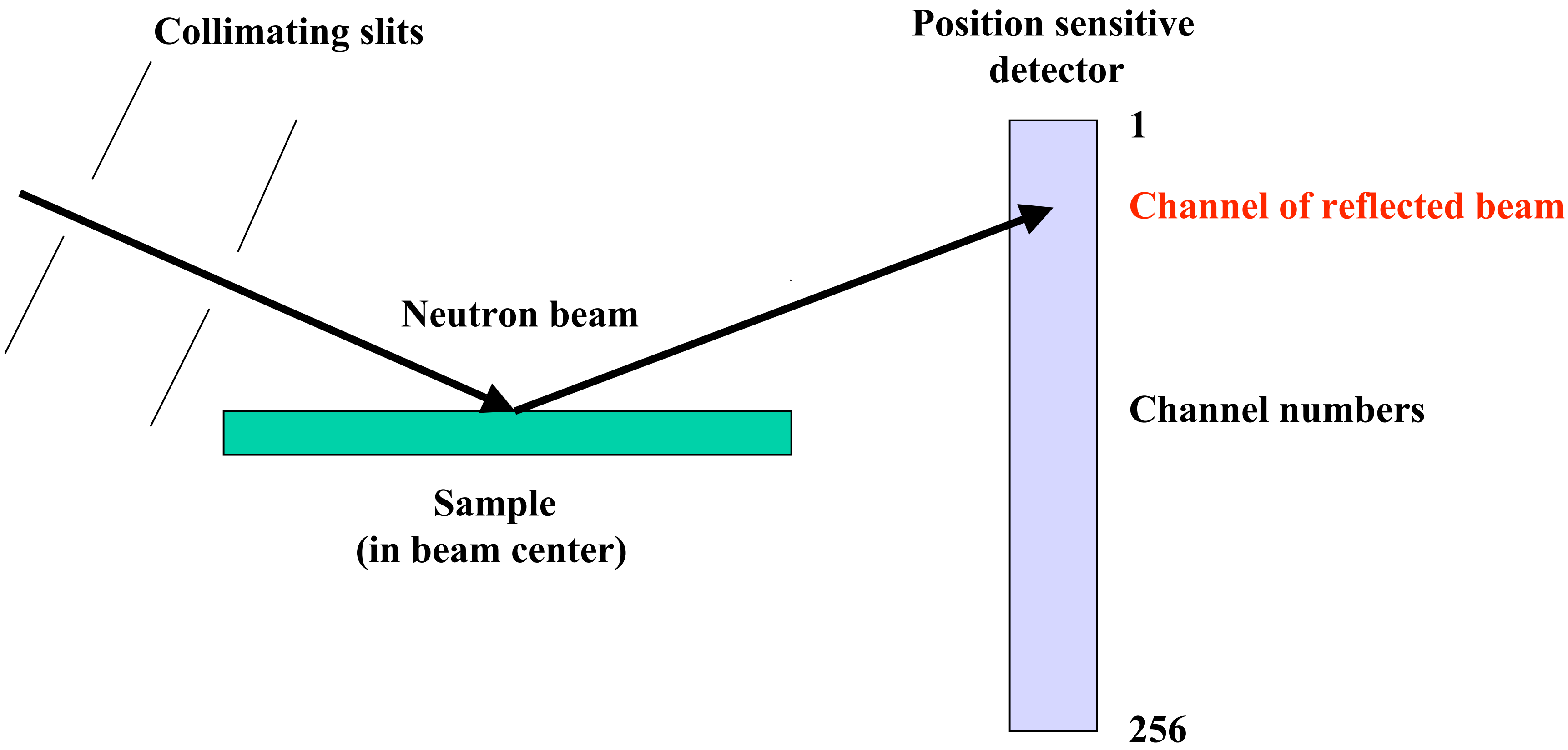
3. Step:
Calculate the reflectivity of the sample

(Divide the "Reflectivity Run" by the "Transmission Run".)

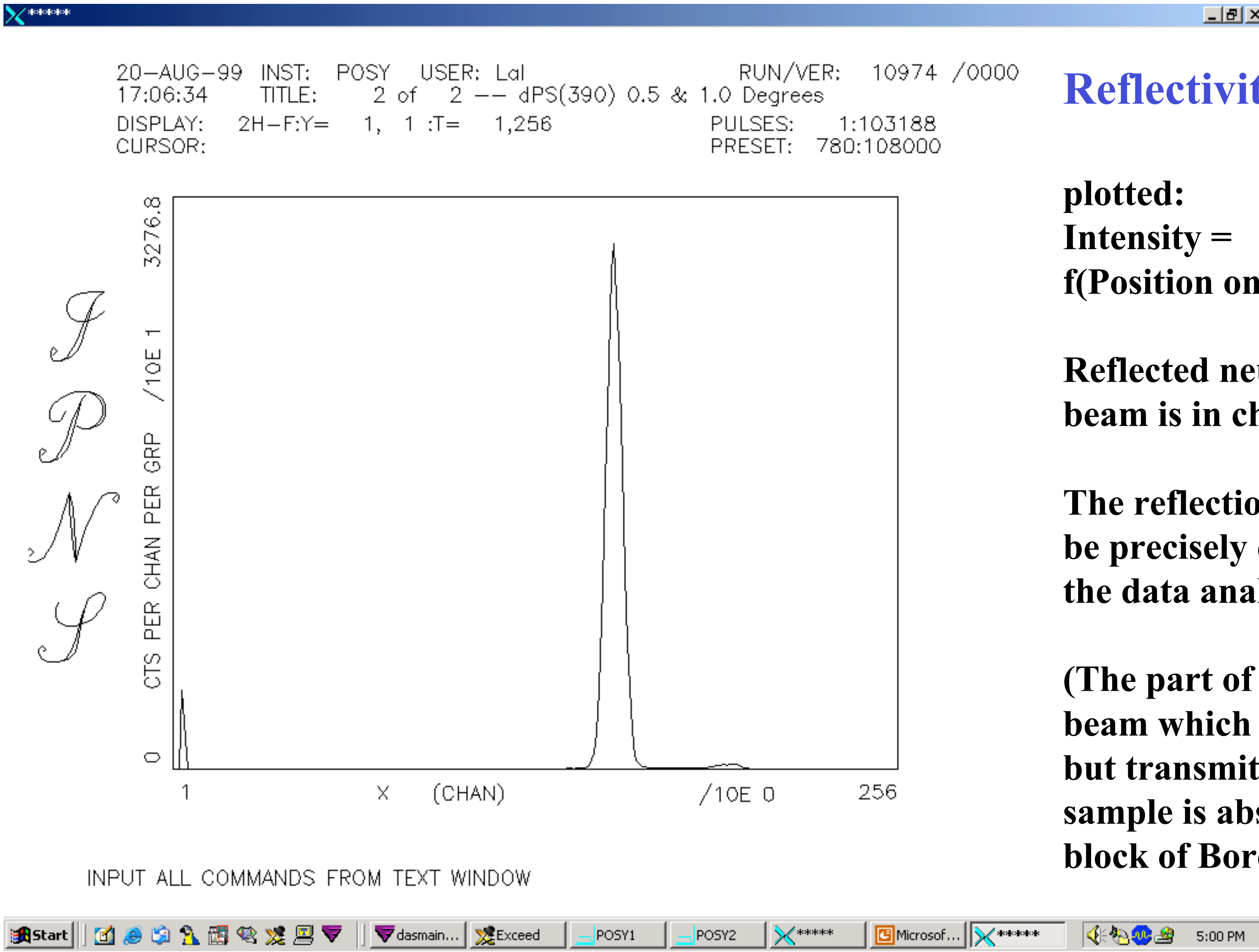


Looking at the Raw Data (POSYII)

Reflectivity run:



Looking at the Raw Data



Reflectivity run:

plotted:
Intensity =
f(Position on detector)

Reflected neutron beam is in channel 156.

The reflection angle will be precisely calculated by the data analysis program.

(The part of the neutron beam which is not reflected but transmitted through the sample is absorbed by a block of Boronnitride.)

4. Step:
Analyze the measured data to learn about your sample

(Use "Trial and Error" method but put as much previous knowledge in your models as you can !
- What are the approximate scattering length densities of your sample ?
- What are the approximate layer thicknesses ?
etc.)

Experiment Schedule

Experiment module	Room	Assistant
Module A Sample preparation/ spin coating	Outside Chem. Lab C248	Xuesong Hu
Module B Monte Carlo simulations	A223	Suzanne te Velthuis
Module C Experiment	POSYII	R.S. Krishnan
Module D Data analysis	A223	Rick Goyette & Abdel Al-Smadi

Experiment Schedule

Time	Group 1	Group 2	Group 3	Group 4
Wed 3:30-5:30 pm	**Module A**	**Module D**	**Module C**	**Module B**
Wed 5:30-7:30 pm	**Module B**	**Module A**	**Module D**	**Module C**
Thu 2:45 -4:45 pm	**Module C**	**Module B**	**Module A**	**Module D**
Thr 4:45 - 6:45 pm	**Module D**	**Module C**	**Module B**	**Module A**

Looking at the Raw Data (POSYI)

POSYI